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October 27, 2004

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Certified by

Jon W Dudas

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PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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INVENTOR(S)

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☐ *Additional inventors are being named on the _____ separately numbered sheets attached hereto*

TITLE OF THE INVENTION (500 characters max)

Process For Preparing High Purity TNT

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ENCLOSED APPLICATION PARTS *(check all that apply)*

☑ Specification *Number of Pages* 7

☐ Drawing(s) *Number of Sheets*

☐ Application Data Sheet. See 37 CFR 1.76

☐ CD(s), Number

☐ Other (specify)

METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☐ Applicant claims small entity status. See 37 CFR 1.27.

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☑ The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number: 190130

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The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

☑ No.

☐ Yes, the name of the U.S. Government agency and the Government contract number are: _____

Respectfully submitted,

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Date 04/21/2003

REGISTRATION NO. *(if appropriate)* 29,404

Docket Number: 20030081 PRO

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This collection of information is required by 37 CFR 1.51. The information is used by the public to file (and by the PTO to process) a provisional application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 8 hours to complete, including gathering, preparing, and submitting the complete provisional application to the PTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, Washington, D.C. 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Box Provisional Application, Assistant Commissioner for Patents, Washington, D.C. 20231.

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EXPRESS MAIL CERTIFICATE EV 195655825 US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: KYLER, et al.

Filed: Herein · Atty. Dkt. No: 20030081 PRO

For: PROCESS FOR PREPARING HIGH PURITY TNT

To: Box Provisional Application & Fee
Commissioner for Patents
Washington, D.C. 20231

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PATENT TRADEMARK OFFICE

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Date: Apr. 21, 2003 Daniel J. Long
Daniel J. Long

Dear Honorable Commissioner:

LETTER OF TRANSMITTAL

Submitted herewith is a Provisional Patent Application consisting of 1 pages of cover sheet, 7 pages of specification and claims.

[ ] Invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

DEPOSIT ACCOUNT **190130** AUTHORIZATION – The Office is hereby authorized to charge the required fee, and any deficiency or credit any overpayment in the fees to the above deposit account, owned by BAE SYSTEMS Information and Electronic Systems Integration Inc.

Respectfully submitted,

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PROCESS FOR PREPARING HIGH PURITY TNT

Background of the Invention

1. Field of the Invention

The present invention relates to organic chemistry and more particularly to the processing of aromatic hydrocarbons. Still more particularly the present invention relates to processes for preparing trinitrotoluene (TNT).

2. Brief Description of Prior Developments

In the prior art process of preparing TNT, one mole of 2,4-dinitrotoluene is added to a mixture of 3 moles of concentrated nitric acid and 5 moles of concentrated sulfuric acid. The mixture is heated to 130° C for 2 hours. The dark red brown viscous solution is poured into a large volume of water and the crude TNT product is isolated by filtration, then purified by washing with sodium sulfite solution which affords a "red water" waste containing TNT isomers and impurities. The TNT is then washed with hot water. The yield of TNT is approximately 84%

A disadvantage of this prior art process is due to the fact that a large volume of concentrated sulfuric acid is used. The acid mixture is highly corrosive. The initial crude TNT is impure and must be purified by sulfite washing which produces a environmentally hazardous waste. The resulting large volume of spent sulfuric acid must be recovery and purified for reuse.

Summary of Invention

An object of the present invention is to overcome the limitations of the prior art methods requiring large quantities of strong oxidizing acids such as sulfuric acid while maintaining an economically useful process.

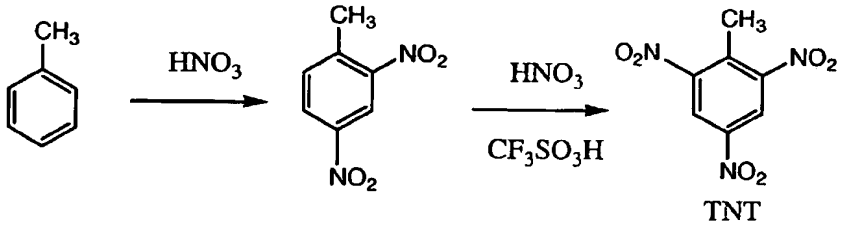
Another object of the present invention is to avoid producing the environmentally hazardous "red water" waste stream which is created with other methods.

In the process of the present invention, TNT is prepared in high purity by either a two step process beginning with toluene or alternatively a one step process starting with dinitrotoluene.

Toluene reacts with 98-99% nitric acid at <30° C to afford a high yield of high purity dinitrotoluene which can be readily converted to TNT by heating this material with a mixture of 98-99% nitric acid in the presence of only one equivalent of the non-oxidizing acid, trifluoromethanesulfonic acid. The process affords very high purity TNT

Detailed Description of the Present Invention

In the present invention TNT is prepared from toluene or 2,2-dinitrotoluene by the following reactions.



The invention is further described with reference to the following example:

Example

Materials: 98% HNO_3 - BAE;

Toluene- Burdick Jackson (purchased 1992, no cat.#, no lot #)

Trifluoromethanesulfonic Acid: Acros, cat. # 169890-500, lot# A016403801

1. A 2 L reactor is charged with 504 mL (8 mol) of 98% nitric acid. The acid is cooled to an internal temperature of 5°C (external jacket temperature set at 0°c).
2. Then 214 mL (2 mol) toluene is added dropwise using an addition funnel at a rate to maintain the internal temperature below 30°C. (the addition takes 45-50 min). The reaction is extremely exothermic with each drop of toluene producing a transient orange-brown color which disappears quickly. Only small amounts of NOx fumes are observed.
3. After complete addition of toluene, the homogeneous mixture is warmed to room temperature and the excess nitric acid and water are removed by distillation under a mild vacuum (50 mmHg) with an external jacket temperature of 105°C.
4. The distillation is continued until all the nitric/water has stopped distilling (requires about 30 min), then the molten DNT is maintained under vacuum at 105°C for an addition 30 min.
5. This material is not isolated but is used for subsequent conversion to TNT. After drying at 105°C for 30 min, the molted DNT is cooled to 50°C prior to the addition of the next reagents.
6. The following experimental is the conversion of DNT to TNT.

7. A solution is prepared in a 2 L Erlenmeyer flask by adding, over a 5 min period, 300 grams, 188 mL (200 mL is satisfactory) (2 mol) of trifluoromethanesulfonic acid (triflic acid) to 1210 mL (20 mol) of 98% nitric acid. There is a slight warming of the solution during the addition of the triflic acid and the mixture warms to about 50°C.

8. This solution is then added in one portion to the cooled DNT in the 2 L reactor.

9. The homogeneous orange solution is then heated to reflux and the progress of the reaction may be monitored by thin layer chromatography.

10. An external jacket temperature of 110°C is used for heating and the internal temperature rises slowly from 85°C to about 92°C during the course of the reaction.

11. Various nitric oxide fumes are observed and must be adequately vented throughout the reaction.

12. The mixture is heated for a total of 4 hrs.

13. Then the excess nitric acid is removed by distillation under a medium vacuum of about 50 mmHg distillation at a temperature of 85°C.

14. When the nitric has finished distilling, the molten TNT separates as a yellow oil on top of the triflic acid/water layer.

15. The reactor is cooled to room temperature with vigorous agitation until the TNT crystallizes as pale yellowish white needles.

16. Then 1 L of water is added and the crystalline TNT is collected by suction filtration on a sintered-glass funnel.

17. The TNT is further washed with an addition 2 L of water, and the product is dried by sucking air through the crystals in the funnel until all of the surface water has been thoroughly removed.

18. The product is then dried in an oven for 48 hrs.

19. The yield of TNT is 415 grams, 91%.

20. Analysis of the product shows the purity is >99%.

It will be appreciated that the process of this invention eliminates the use of sulfuric acid by using a less caustic and less oxidizing acid, namely trifluoromethanesulfonic acid (triflic acid). Less acid, only one molar equivalent, is required in the new method, and the temperature of the reaction is lower (85-90°C). A higher purity TNT product is obtain eliminating the sulfite washing stage thus eliminating the hazardous "red water" waste stream.

While the present invention has been described in connection with the preferred embodiments of the various figures, it is to be understood that other similar embodiments may be used or modifications and additions may be made to the described embodiment for performing the same function of the present invention without deviating therefrom. Therefore, the present invention should not be limited to any single embodiment, but rather construed in breadth and scope in accordance with the recitation of the appended claims.

Claims

What is claimed is:

1. A process for preparing trinitrololuene (TNT) comprising the steps of:

   (a) treating toluene with nitric acid having a concentration of about 98% to about 99% by weight at a temperature of less than about 30° C to produce high purity dinitrotoluene; and

   (b) then treating the dinitrotoluene formed in step (b) with nitric acid and trifluoromcthane sulfuric acid to produce TNT.

Abstract

A process for preparing trinitrololuene (TNT) in which toluene is treated with nitric acid having a concentration of about 98% to about 99% by weight at a temperature of less than about 30° C to produce high purity dinitrotoluene. The resulting dinitrotoluene is then treated with nitric acid and trifluoromethane sulfuric acid to produce TNT.

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